

18th International Vacuum CongressExcitation dependence of photoluminescence in the 1.5-1.6 μm wavelength region from grown dislocation-rich Si layersA. A. Shklyayev,^{a,b} A. V. Latyshev^{a,b} and M. Ichikawa^c^a The Institute of Semiconductor Physics SB RAS, Novosibirsk 630090, Russia^b Novosibirsk State University, Novosibirsk 630090, Russia^c Department of Applied Physics, The University of Tokyo, Bunkyo-ku, Tokyo 113-8656, Japan

Abstract

We study photoluminescence (PL) in the 1.5-1.6 μm wavelength region from dislocation-rich Si layers grown using the oxidized Si surfaces. The obtained excitation dependences of the PL intensities are interpreted on the base of a model of the electronic structure that consists of the shallow dislocation-induced conduction sub-band and deep levels located about 0.3 eV above the valence band edge. At low temperature and low excitation conditions the dependences are described well by mean of the Shockley-Read-Hall statistics for the carrier recombination via deep states. The PL data at high excitation conditions suggest that the dominating process that limits the increase in the excess carrier concentration is Auger recombination in which one of three carriers involved into the recombination event is from the deep states. A blue shift of the dislocation-related PL peak with increasing excitation intensity is observed, which is associated with changes in the occupancy of the deep states.

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1. Introduction

Infrared emitters for the 1.5-1.6 μm wavelength region can be fabricated from silicon with dislocation-induced deep levels [1-5]. Dislocations are usually introduced into already existing Si crystals. Recently, we developed a method for the *growth* of Si layers with a high concentration of dislocations, which exhibit only one dislocation-related photoluminescence (PL) peak [6,7]. The growth is realized on the nanostructured surface composed of dense arrays of Ge islands which are formed when germanium is deposited on the oxidized Si surface [8].

In this work we measure the excitation dependences of the PL from the grown dislocation-rich Si layers and consider the dependences using the Shockley-Read-Hall (SRH) statistics [9-11] for the recombination processes via dislocation-related deep states. The obtained results show that radiative and nonradiative carrier recombination via deep states dominates over other recombination processes only at low temperatures of about 4 K and at relatively low excitation conditions. At high excitation conditions Auger recombination that involves carriers from the deep states are prevailing. It is found that the dislocation-related PL peak is shifted towards the shorter wavelength region as the pump power density increases. The shift is associated with changes in occupancy of the deep levels. The

superlinear dependences of the dislocation-related PL intensity on the pump power density, which are observed at 100 and 170 K, probably reflect the interaction between the deep states. This can occur due to the temperature-induced delocalization of the deep states and their high concentration.

2. Experimental

The dislocation-rich Si layers were grown by means of molecular beam epitaxy on *p*-type FZ-grown Si(100) wafers with a resistivity of 20-100 Ω cm. The preparation of the initial surface for the growth included the deposition of a buffer layer about 100 nm thick followed by oxidation of its surface to form a 0.3-0.5 nm thick Si oxide film. The oxidized Si surface was then covered with about 1.0 nm of Ge to produce nanoscale Ge islands with a density of their arrays of about 2×10^{12} cm⁻². The formation of the dense arrays of Ge islands was well reproducible in a wide range of parameters such as growth temperature, deposition rate, and coverage. The subsequent deposition of silicon was performed at temperatures of about 450 °C at which a high density of stacking faults and dislocations are introduced in the growing Si layer [7]. The details of the growth mechanism were described elsewhere [12,13].

Morphology of the surfaces after different stages of the growth was investigated using a scanning tunneling microscope (STM). The PL data were mostly obtained for the dislocated Si layers about 100 nm thick. PL spectra were measured using a standard lock-in technique in conjunction with an InGaAs photomultiplier detector or a Ge detector, a grating monochromator, and the 532 nm line of a frequency-doubled diode-pumped Nd:YAG laser. The maximum laser power was about 80 mW with the beam diameter of ~ 0.3 mm on the sample surface. Additionally, the 325 nm line of a He–Cd laser was used in some test experiments.

3. Morphology of the grown surfaces

The STM study showed that the surface of the clean Si substrate before the oxidation was atomically flat and consisted of atomic steps distributed uniformly along the surface. The STM images of the oxidized Si surface contained randomly distributed bright points which evidence for the existence of leakages of the tunneling current through the oxidized Si film rather than for nonuniformity of the surface morphology. The deposition of germanium on the oxidized Si surface leads to the formation of dense arrays of germanium islands (Figure 1). It has been shown that at growth temperatures from about 430 to 550 °C the array of Ge islands is composed of crystalline Ge islands with a part of them grown epitaxially with respect to the Si substrate [8].

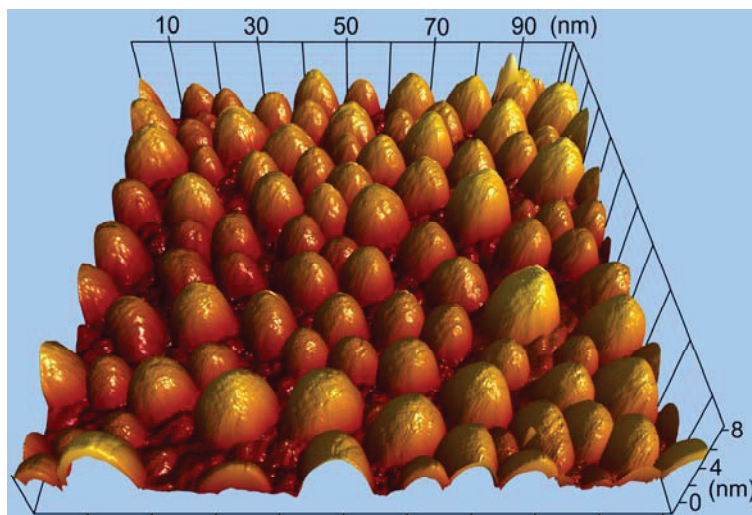


Fig.1 STM image of the dense array of germanium islands which appear after deposition of about 1 nm of Ge on the oxidized Si surface at 450 °C.

The deposition of silicon on such a nanostructured surface at temperatures from about 400 to 500 °C results in the formation of a Si layer with a high concentration of dislocations, so that the distance between the dislocations can be as small as 10 nm. We found that at these growth temperatures the morphology of the growing Si surface is not atomically flat. Silicon growth on the Ge islands occurs by mean of the attachment of Si atoms to the surface of Ge islands, but not to areas of the oxidized Si surface between them [12]. These mechanism leads to the formation of dislocations in the places of coalescence of growing islands and also to the development of rough surface morphology, as shown in Figure 2. One of the possible reasons for the development of such morphology is the presence of lattice strain around the dislocation core which creates a potential barrier to the attachment of deposited atoms. Since the crystal defect that is expected to produce a deep state in the Si band-gap is the stable interstitial cluster bound to the dislocation core, the density of the deep states can be estimated as $\sim 1 \times 10^{18} \text{ cm}^{-3}$, corresponding to about one state per ten spacings along the dislocation line [14] with the concentration of dislocations of the order of 10^{11} - 10^{12} cm^{-2} .

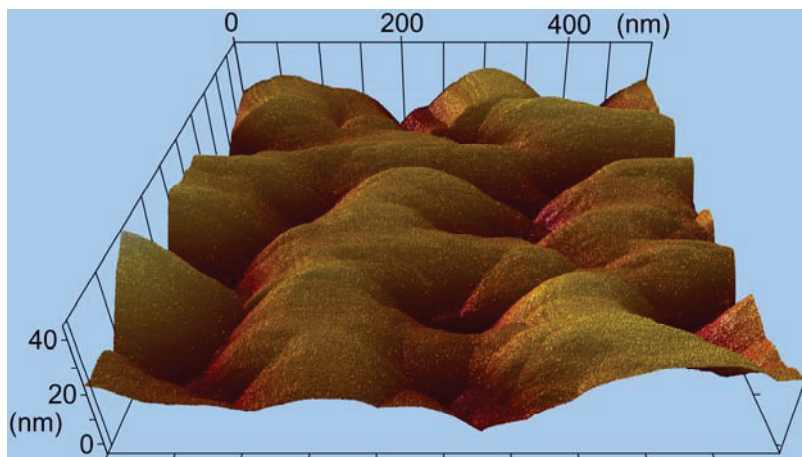


Fig.2 STM image of the Si surface which forms after deposition of about 50 nm of silicon on the dense array of Ge islands at 450 °C.

4. PL of the grown Si layers

The Si layers grown in the temperature range from 400 to 500 °C produce PL in the broad wavelength region from 1.2 to 1.7 μm [6,13] due to the presence of Ge quantum dots and different crystal defects. Annealing of the sample at 900 °C leads to the appearance of an intense PL peak in the 1.5-1.6 μm wavelength region (Figure 3), whereas the PL at the other wavelengths substantially decreases. It has been shown that after high-temperature annealing the PL peak centered near 1.55 μm does not relate to carrier recombination via electronic states in the Ge quantum dots because Ge dot-related PL disappears after annealing of the samples at temperatures above 750 °C [6,15]. The position of the PL peak and the correlation of its intensity with the concentration of dislocations pointed out that the PL originates from crystal defects in the grown Si layer. Since dislocated Si samples prepared by different methods usually produce several defect-related PL peaks, the observation of only one PL peak near 1.55 μm indicates that our growth and annealing procedure produce Si layers with only one type of radiative defect-induced deep states.

A scheme of energy levels in the dislocated silicon, which would be associated with the PL peak in the 1.5-1.6 μm region, has not yet been finally established. Defects that form the dislocation cores do not produce deep energy levels in the Si band-gap according to both theoretical calculations [16,17] and experimental results [18]. However, they can create the shallow sub-bands which are located at levels close to the conduction and valence band edges [17-22]. At the same time, it has been shown in the theoretical calculations that the defects such as stable interstitial clusters bound to the dislocation core produce levels about 0.3-0.4 eV above the valence band edge (E_v) [14]. The

existence of such levels in dislocated silicon is also followed from the different experiments [18,22,23]. Therefore, the dislocation-related PL is suggested to be the result of electron transitions between the conduction sub-band and the deep states located near $E_v + 0.3$ eV (Figure 3). The similar scheme of energy levels was recently considered by Yu et al. [24] to explain the origin of the electroluminescence spectrum with only one peak centered near 1.5 μm from silicon with the dislocation network.

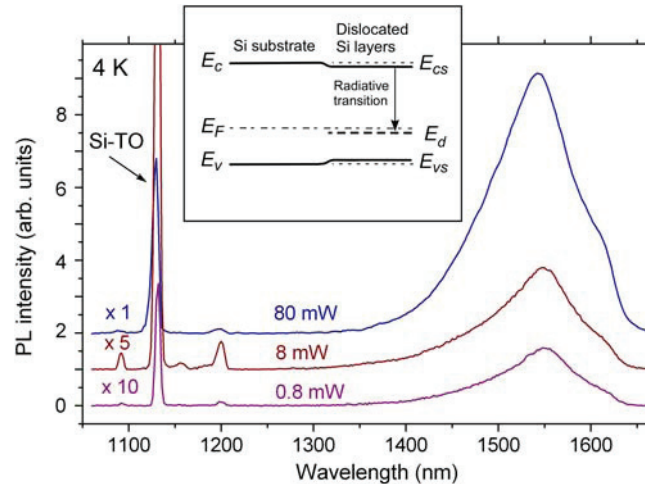


Fig.3 Low-temperature PL spectra from grown dislocation-rich Si layers. The spectra contain peaks of phonon-assisted interband transitions in the crystalline Si substrate with the most intense one marked as Si-TO. Inset shows the schematic diagram of the band structure of the dislocated silicon. Here E_c , E_F , E_{cs} , E_{vs} and E_d are respectively the energies of the conduction band edge, Fermi level, conduction and valence sub-band edges and deep states.

5. Excitation dependence of the intensity of PL peaks

The relation between the intensities of Si-TO peak from the Si substrate and the dislocation-related peak from the grown Si layer in the PL spectrum from the sample depends on the thickness of the layer and the wavelength of the excitation laser beam. The PL spectra shown in Figure 3 for the sample with the 100 nm thick grown layer of dislocated silicon were measured using the 532 nm wavelength excitation laser beam which is characterized by a relatively large penetration depth of ~ 1 μm for silicon. However, when the 325 nm laser beam with the much smaller penetration depth of the order of 10 nm is used for the excitation, the intensity of the Si-TO peak becomes significantly lower than the intensity of the dislocation-related peak. Moreover, at such an excitation wavelength the dislocation-related peak completely disappears in PL spectra from samples with the 300 nm thick dislocated Si layers. This result indicates that all the excess carriers recombine in the dislocated Si layer and do not reach the Si substrate by diffusion. This means that the carrier recombination in the dislocated Si layers entirely occurs with the assistance of dislocation-induced states. Therefore, in the further analysis it is accepted that the Si-TO peak in PL spectra from samples with the dislocated Si layers exclusively originates from the Si substrate.

We measured the dislocation-related PL peak intensity (I_{PL}) as a function of the excitation power density (W) for several temperatures. Excitation dependences of I_{PL} are usually approximated by a power law, $I_{PL}(W) \sim W^m$, where m is the power exponent, as shown in Figure 4. The obtained dependences show that the value of m depends on the temperature and the range of W . The values of m were obtained for several samples and were essentially the same for the used Ge and InGaAs-photomultiplier detectors. The results show that the dependence $I_{PL}(W)$ of the Si-TO peak at 4 K is characterized by the greater values of m than the defect-related PL peak. However, the increase in the defect-related PL peak intensity with increasing W is observed in the whole range of W used, whereas the

intensity of the Si-TO peak reaches a maximum and then shows the tendency to decreasing [Figure 4(a)]. The other feature which should be mentioned is that the value of m for the defect-related PL peak increases with temperature.

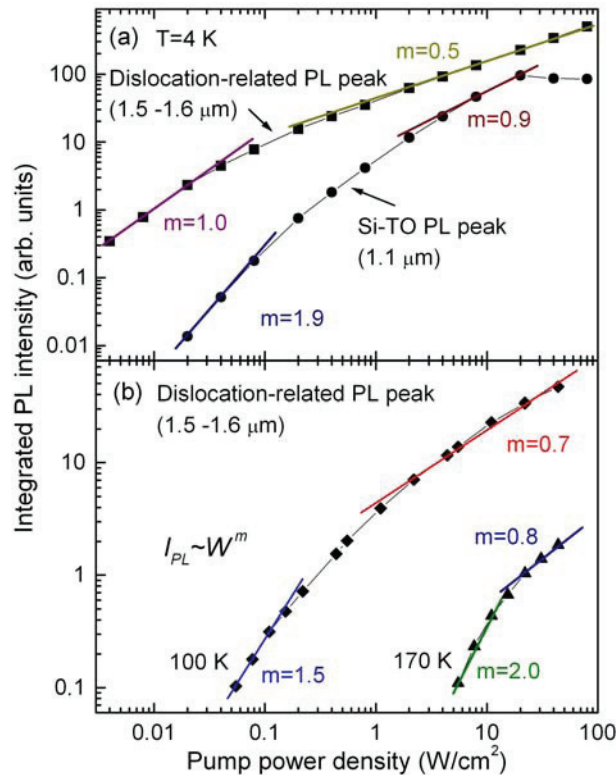


Fig.4 (a) Intensities of the Si-TO and dislocation-related PL peaks as a function of the pump power density at 4 K. (b) Dependence of the intensity of the dislocation-related PL peak (around 1.55 μm) on the pump power density for two temperatures of 100 and 170 K. The lines show the approximation of the experimental data by the power-law function. Values of the power exponent m are indicated in the figure at the corresponding approximation lines.

6. PL of crystalline silicon

In spite of the commonly accepted point of view that the probability of interband radiative transitions in silicon is very low due to the indirect band structure, it was recently shown that the intrinsic quantum yield of luminescence can reach 20 % [25]. Such a possibility is suggested to be realized in the float-zone silicon in which the concentration of nonradiative recombination centers induced by impurities and crystal defects is essentially reduced. The other case of elevated radiative efficiency was observed in the structures prepared by the ion implantation of boron which leads to the formation of doped silicon with a number of dislocation loops [26,27]. The authors suggest that the enhancement of luminescence occurs due to carrier localization near the dislocations, which takes place under the dislocation-induced strain field. The external quantum yield of luminescence in this case was estimated to be about 0.1-1 %, depending on the configuration of emitting structures. It can be also mentioned that the carrier localization in the Si clusters surrounded by SiO_x also leads to high values of quantum yield of luminescence [28]. In the commercial float-zone silicon of which our Si substrates were made, the internal quantum yield of luminescence is expected to be below 1 %. In the further analysis we suggest that the concentration of free carriers under the steady-state conditions is governed by the balance between the photo- and thermogeneration rates from

one side and the nonradiative recombination rates from the other side, whereas the influence of the radiative recombination is negligible.

At relatively low values of W , the rate of photoexcitation $R(W) = \sigma_{ex}W$, where σ_{ex} is the excitation cross section, is mostly balanced by the rate of nonradiative SRH (Shockley-Read-Hall) recombination, which occurs via deep recombination centers. The SRH recombination rate is given by [9-11]

$$R_{SRH}(n, p) \approx c_n c_p N_r (np - n_i^2) / [c_n(n + n_1) + c_p(p + p_1)], \quad (1)$$

where n and p are the concentrations of free electrons and holes, c_n and c_p are the probability that electrons and holes will be captured by empty and occupied centers of nonradiative recombination, respectively, N_r is the concentration of nonradiative recombination centers, n_i is the intrinsic carrier concentration, $n_1 = N_c \exp[-(E_c - E_r)/kT]$ and $p_1 = N_v \exp[-(E_r - E_v)/kT]$. Here E_c , E_v and E_r are energy levels of the conduction and valence band edges and the nonradiative recombination centers, and N_c and N_v are the density of states in the conduction and valence bands, respectively.

In the conditions when the nonradiative recombination dominates, the probabilities c_n and c_p are about equal [29]. The PL from silicon becomes detectable at relatively high excitation conditions at which $n \approx \Delta n$, $p \approx \Delta p$ and $p \approx n$ are the reasonable approximation for low temperatures, where Δn and Δp are the concentrations of excess electrons and holes generated by the photoexcitation, respectively. At low temperatures we also have n_1 and $p_1 \ll n$, and $n_i \ll n$. Equation (1) is transformed into the form

$$R_{SRH}(n, p) \approx c_n N_r n / 2. \quad (2)$$

Taking into account that $R_{SRH}(n, p) \approx \sigma_{ex}W$, one can obtain $n \approx 2\sigma_{ex}W / c_n N_r$. Then, the radiative recombination rate of free carriers is $I_{PL}(W) = Bnp \sim W^2$, where B is the rate coefficient [29]. The derived relationship is in good agreement with the experimental data obtained for the Si-TO peak in the range of relatively small values of W at which the approximation of the experimental dependence by the power law is characterized by the power exponent $m \approx 1.9$ [Figure 4(a)].

As the value of W increases, Auger processes become dominating, so that

$$\sigma_{ex}W \approx C_1 n^2 p + C_2 n p^2, \quad (3)$$

where C_1 and C_2 are the coefficients of Auger recombination rates. Since n and p have the same dependence on W , Equation (3) leads to n and $p \sim W^{1/3}$, and thus $I_{PL}(W) \sim W^{2/3}$. The tendency to the decrease in the value of m with increasing W is in agreement with the obtained experimental dependence [Figure 4(a)].

In the range of high excitation conditions, the photon absorption by free carriers becomes essential and leads to the decrease in the concentration of electron-hole pairs which are responsible for the radiative recombination. This causes the decrease in the intensity of the Si-TO peak in spite of the increase in the pump power density [Figure 4(a)].

7. Recombination processes in the dislocated Si layers

We can assume that in the dislocation-rich Si layers the radiative and nonradiative recombination occurs via the same type of deep states located about 0.3-0.4 eV above the valence band edge. According to SRH statistics, the equation similar to Equation (1) for the transition rate of electrons from the dislocation-induced conduction sub-band to the deep levels can be written as

$$R_{SRH}(n_s, p_s) = c_{cd} c_{dv} N_d (n_s p_s - n_i^2) / [c_{cd}(n_s + n_{1s}) + c_{dv}(p_s + p_{1s})], \quad (4)$$

where n_s and p_s are the concentrations of free electrons and holes in the conduction and valence dislocation-induced sub-bands, respectively, c_{cd} and c_{dv} are the probabilities of electron transitions from the conduction sub-band to an empty deep state and from the deep level to the valence sub-band, N_d is the concentration of deep states, $n_{1s} = N_{cs} \exp[-(E_{cs} - E_r)/kT]$ and $p_{1s} = N_{vs} \exp[-(E_r - E_{vs})/kT]$. Here E_{cs} , E_{vs} and E_d are energy levels of the conduction and valence sub-band edges and the deep levels, and N_{cs} and N_{vs} are the density of states in the conduction and valence sub-bands, respectively. Since a part of the transitions are radiative, the intensity of dislocation-related PL peak is proportional to the transition rate, $I_{PL}(n_s, p_s) = \delta R_{SRH}(n_s, p_s) = \delta \sigma_{ex}W$, where δ

is the coefficient of proportionality. The linear dependence of I_{PL} on W is observed at the low temperature and at relatively low excitation conditions [Figure 4(a)].

At low temperature, according to the scheme of energy levels (Figure 3), the deep states are almost fully occupied, so that the electron transitions are limited by the stage of trapping of holes in the deep states. At high excitation conditions, Auger recombination becomes dominating over the SRH recombination, leading to the balance of the rates in the form

$$\sigma_{ex}W \approx C_1 n_s^2 p_s + C_2 n_s p_s^2 + C_3 f N_d n_s p_s + C_4 (1-f) N_d n_s p_s, \quad (5)$$

where C_i is the rate coefficient of the corresponding Auger process and f is the probability that the deep state is occupied. If assume that equation $R_{SRH}(n_s, p_s) \approx c_n N_d p_s / 2$ which is the similar to Equation (2) is applicable for describing the radiative transitions via deep states in spite of the fact that Auger recombination has not been taken into account in the simple SRH statistics, the experimental dependence with $m \approx 0.5$ [Figure 4(a)] can be realized when the third and fourth terms in Equation (5) dominates over others, so that

$$I_{PL} \sim p_s \approx \{\sigma_{ex}W / [C_3 f N_d + C_4 (1-f) N_d]\}^{1/2}. \quad (6)$$

This means that one of three carriers that are involved into a dominating Auger recombination event is from the deep states. This is reasonable for the Si layers with a high concentration of deep states.

At larger temperatures the dislocation-related PL becomes detectable at substantially higher level of the excitation power density [Figure 4(b)]. Such behavior occurs due to the strong temperature-induced increase in the rate coefficients of carrier generation whereas the rate coefficients of carrier recombination have weak dependences on temperature. This leads to the increase in the carrier concentration with temperature, so that the influence of the photogeneration on the carrier concentration remains insufficient up to large values of excitation intensities. The values of the power exponent larger than $m=1$ obtained for the excitation dependences [Figure 4(b)] indicate that the simple SRH statistics does not applicable at temperatures of 100 and 170 K. This can arise from a high concentration of deep states. The increase in temperature causes the delocalization of the deep states leading to their overlapping. The deep states may thus create a sub-band in which the trapped carriers possess properties of free carriers. The value of the power exponent up to $m \approx 2$ is expected for the radiative recombination of electron-hole pairs ($I_{PL} = Bnp$) [30], when the carrier densities are governed by the nonradiative SRH recombination.

8. Blue shift of the dislocation-related PL peak

Figure 5 shows that the position of the dislocation-related PL peak gradually shifts towards the short-wave region, as the excitation power density increases. The shift is not the result of heating the sample with the laser beam because the heat leads to the decrease of the Si band-gap and causes the shift of PL peaks towards the long-wave region. Figure 6 shows that the dependence of the PL peak position on the excitation power density is different at 5 and 100 K. Since the PL peak position is determined by the occupancy of the deep states, the observed shift suggests that the occupancy depends on the excitation intensity.

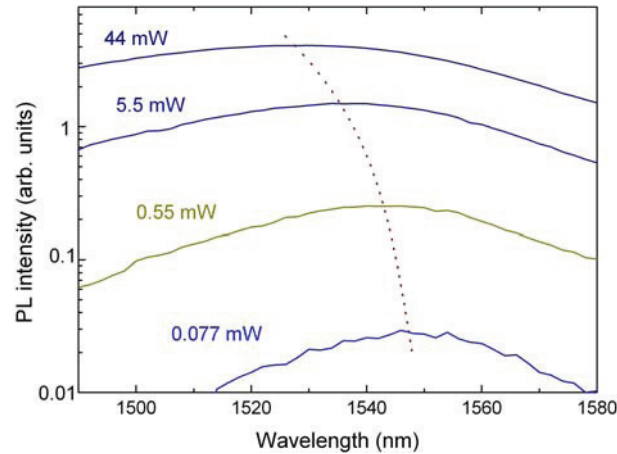


Fig.5 Dislocation-related PL peak at different pump power densities at 100 K. The dotted line shows positions of the maximum PL peak intensity.

The shift of the PL peak at 4 K is observed at high excitation conditions at which the Auger recombination dominates, as is shown in the previous section. In order to correctly describe the excitation dependence of the occupancy of deep states, the simple SRH statistics have to be modified by means of accounting the Auger recombination in the balance between carrier generation and recombination processes.

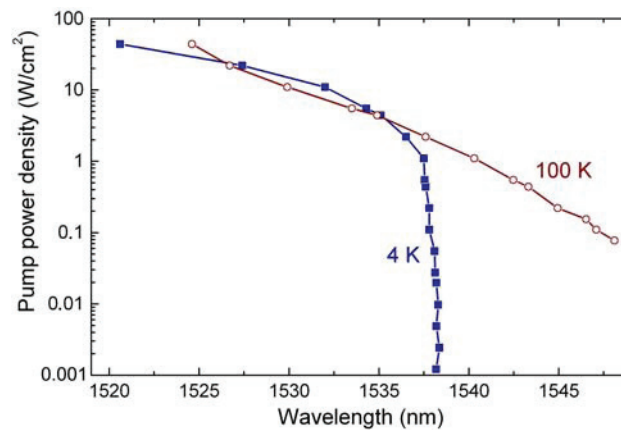


Fig.6 Dependence of the dislocation-related PL peak position versus the pump power density at 4 and 100 K.

In conclusion, it can be mentioned that diodes fabricated on the base of grown dislocation-rich Si layers exhibit light emission in the 1.4-1.6 μm wavelength region at room temperature [5]. The increase of the light emission efficiency of the diodes can be achieved by utilizing the properties of optical microcavities. When microcavities are formed in an optically active layer, they strongly influence on its electromagnetic properties. Due to optical resonance, light emissions at particular discrete wavelengths are selected and enhanced [31]. For the activation of the microcavity features in full measure, the layers have to be prepared for a given thickness in the silicon-on-insulator configuration. It seems that only our growth method at present is capable producing such structures with dislocation-rich Si layers.

9. Summary

We use the nanostructured surface composed of dense arrays of Ge islands prepared by Ge deposition on the oxidized Si surface to grow dislocation-rich Si layers. The grown Si layers exhibit intense PL peak centered in the

1.5-1.6 wavelength region after high-temperature annealing. According to the probable model of electronic structure of dislocated silicon, we assume that the radiative transitions in the grown dislocation-rich Si layers occur between the dislocation-induced conduction sub-band and deep levels located about 0.3 eV above the valence band edge. The excitation dependences of the PL peak intensity are considered involving the SRH statistics for the transitions via deep states, and Auger recombination. The SRH statistics describes well the dependences at low temperature and low excitation conditions. At high excitation conditions, Auger recombination that involves carriers from the deep states plays an essential role in determining the carrier densities. The observed blue-shift of the dislocation-related PL peak is associated with the change in the occupancy of the deep states.

Acknowledgments

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